

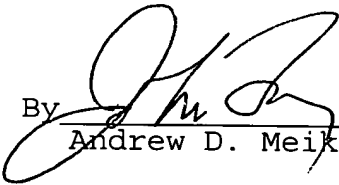
- ☒ An extension of two (2) month(s) was previously requested and paid for on October 30, 2003 in the instant application. Thus, no extension fee is required.
- ☒ Fee for filing an Appeal Brief - \$330.00 (large entity).
- ☒ Check(s) in the amount of \$330.00 is(are) attached.
- ☐ Please charge Deposit Account No. 02-2448 in the amount of \$0.00. A triplicate copy of this sheet is attached.

Appl. No. 09/194,112

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. §§ 1.16 or 1.17; particularly, extension of time fees.

Respectfully submitted,

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(Rev. 11/04/03)



PATENT
0229-0532P

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant: Masahide Onuki et al. Conf.: 2812
Appl. No.: 09/194,112 Group: 3711
Filed: November 23, 1998 Examiner: Blau, S..
For: GOLF CLUB HEAD

APPEAL BRIEF

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Assistant Commissioner for Patents
Washington, DC 20231

November 7, 2003

Sir:

In response to the Examiner's Advisory Action dated May 27, 2003, and the Examiner's Final Office Action dated January 7, 2003, the following Appeal Brief is respectfully submitted in connection with the above-identified application.

11/10/2003 JBALINAM 00000045 09194112

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I. Real Party in Interest

The real party in interest of the present invention is Sumitomo Rubber Industries, Ltd. Of Hyogo-Ken, Japan, the assignee of the entire right and interest of the instant application. The assignment of said right and interest was recorded on November 23, 1998 at Reel 9768, Frame 0275.

II. Related Appeals and Interferences

There are no related appeals or interferences pending for the present application.

III. Status of Claims

Claims 1-50 are pending in the present application. Claims 39-41 and claims 48-50 have been withdrawn from a prior election of species requirement.

Claims 21, 23-25 and 30 are rejected under 35 USC §103(a) as being unpatentable over Peker '642 (US Patent No. 5,896,642).

Claims 1-20, 22, 26-29, and 31-32 are rejected under 35 USC §103(a) as being unpatentable over Peker '642 in view of Kobayashi '742 (US Patent No. 5,611,742).

Claims 42-43 are rejected under 35 USC §103(a) as being unpatentable over Peker '642 in view of Kobayashi '742 and further in view of Anderson '663 (US Patent No. 5,261,663).

Claims 1, 5, and 45-46 are rejected under 35 USC §103(a) as being unpatentable over Peker '642 in view of Kobayashi '501 (US Patent No. 5,601,501) and further in view of Sieleman '005 (US Patent No. 5,792,005).

Thus, the rejections concerning claims 1-38 and 42-46 are appealed.

IV. Status of Amendments

Subsequent to the Examiner's final rejection of claims 1-38 and 42-47 on January 7, 2003, an after-final response was filed May 7, 2003. An attempt was made to amend claim 47 in the after final response of May 7, 2003, but the Examiner denied entry of the amendment.¹ In the reply of May 7, 2003, an attempt was made to change the dependency of claim 47 from 1 to 21. The Examiner denied entry saying that entry of the amendment would require further consideration and/or searching. A Notice of Appeal was filed on July 7, 2003. On October 30, 2003, claim 47 was canceled.

V. Summary of Invention

The present invention, as recited in claim 1, relates to a golf club head comprising a hitting face for golf balls (page 3, lines 17-18), said hitting face formed at least partially by a

metallic material (page 3, line 24), and said metallic material satisfying the following relation:

$$y \geq 0.006x + 60 \quad (\text{page 3, line 21})$$

wherein

x is Young's modulus in units of kgf/mm², and

y is tensile strength in units of kgf/mm², and (page 3, lines 22-23)

wherein said metallic material has a young's modulus of 3,000 to 12,000 kgf/mm² (page 9, lines 3-11), and a tensile strength of 105 to 175 kgf/mm² (page 8, line 26 and Example 3 on page 23) and said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm (page 7, line 13).

VI. Issues to be considered

Issue 1

Are claims 21, 23-25 and 30 patentable over Peker '642 (US Patent No. 5,896,642)?

Issue 2

Are claims 1-20, 22, 26-29, and 31-32 patentable over Peker '642 in view of Kobayashi '742 (US Patent No. 5,611,742)?

¹ The Appendix cites the claims as appealed.

Issue 3

Are claims 42-43 patentable over Peker '642 in view of Kobayashi '742 and further in view of Anderson '663 (US Patent No. 5,261,663)?

Issue 4

Are claims 1, 5, and 45-46 patentable over Peker '642 in view of Kobayashi '501 (US Patent No. 5,601,501) and further in view of Sieleman '005 (US Patent No. 5,792,005)?

VII. Grouping of Claims

Appellants respectfully request that the claims be grouped as follows.

Group I - claims 1-4, 8-10, 14, 16, 18, 27, 33, 36, 42, and 45

Group II - claims 5-7, 11-13, 15, 17, 19, 20, 26, 28, 29, 34, 37, 43, and 46

Group III - claims 21-25, 30-32, 35, 38, and 44

Each group of claims raises different issues for consideration by the honorable Board of Patent Appeals and Interferences as follows:

Group I -Issues 2, 3 and 4

Group II - Issues 2, 3, and 4

Group III - Issues 1, 2 and 4

VIII. Arguments

Argument 1

Claims 21, 23-25 and 30 are patentable over Peker '642 (US Patent No. 5,896,642)

Appellants assert that the Examiner has failed to make out a *prima facie* case of obviousness with regard to the 35 USC §103(a) rejection of claims 21, 23-25 and 30 over Peker '642. Three criteria must be met to make out a *prima facie* case of obviousness.

- 1) There must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings.
- 2) There must be a reasonable expectation of success.
- 3) The prior art reference (or references when combined) must teach or suggest all the claim limitations.

See MPEP §2142 and *In re Vaeck*, 20 USPQ2d 1438 (Fed. Cir. 1991).

The Examiner has failed to meet any of these elements to make a *prima facie* obviousness rejection.

Claim 21 recites:

A golf club head comprising a hitting face for golf balls, said hitting face formed at least partially by a metallic material, and said metallic material satisfying the following relationship:

$$y \geq 0.006x + 60$$

wherein

x is Young's modulus in units of kgf/mm^2 , and

y is tensile strength in units kgf/mm^2 , and

wherein said metallic material has a Young's modulus of 5,000 to 16,000 kgf/mm² and a tensile strength of 105 to 175 kgf/mm².

Appellants assert that the Examiner has failed to show that Peker '642 discloses or suggests all of the elements of the above claim 1. Attached to this appeal brief, please find a 37 CFR §1.132 declaration (Appendix B) filed with the Reply of May 7, 2003 with a Table demonstrating some of the elements claimed in claim 21 and where some of them can be found or not found in Peker '642. The Table is reproduced for the reader's benefit here.

Claimed Invention Claim 21	Disclosure of Peker '642
x is Young's modulus in units of kgf/mm ² and is 5,000 to 16,000 kgf/mm ²	No disclosure in Peker '642
y is tensile strength in units kgf/mm ² and is 105 to 175 kgf/mm ²	Peker '642 discloses a tensile strength of 194 kgf/mm ²

Regarding the above Table, one should note that Peker '642 does not disclose or suggest a Young's modulus. The tensile strength of the claimed invention is 105 to 175 kgf/mm² whereas Peker '642 only discloses a tensile strength of 194 kgf/mm² (please see column 4, lines 28-29 in Peker '642 - to convert from gigaPascal to kgf/mm² divide by 9.80665×10^{-3}). Please note that the tensile strength disclosed in Peker '642 is outside of the claimed range. Accordingly, Appellants assert that Peker '642 does not teach or

suggest all of the claimed limitations and thus, for this reason alone, has failed to make out a *prima facie* case using Peker '642.

The Examiner in the Office Action of January 7, 2003 states:

Peker discloses a golf club head having a hitting face formed of a metallic amorphous metal, an amorphous alloy of zirconium base, Ni, Al, Cu, Hf, tensile strength 1.9 Gpa (194 kgf/mm²) (Col.4 Lns. 12-43), a material meeting the formula M_aX_b with $65 \leq a \leq 100$ and $0 \leq b \leq 35$ in the form of M(Zr/Ti/Be/Cu/N) of 100 (Col. 4, Lns. 13-16), a material meeting the formula $Zr_cM_dX_e$ with $20 \leq d \leq 80$, and $0 \leq e \leq 35$ in the form of Zr of 41.2, M(Ti/Be/Cu/Ni) of 58.8, and X(Hf) of 0 (Col.4, Lns. 23-27), and material meeting the formula $Zr_cM_dX_e$ with $50 \leq c \leq 75$, $25 \leq d \leq 50$, $0 \leq e \leq 1$ in the form of Zr of 60, M(Al/Ni) of 40 and X(Hf) of 0 (Col. 4, Lns. 39-43) and an iron head (Fig. 6). Clearly the hitting face material has a Young's modulus and a hardness and one skilled in the art in manufacturing a hard face with mixtures of elements of Zr/Ti/Be/Cu/Ni/Hf would have selected a composition having a suitable Young's modulus and tensile strength in which Young's modulus of 5,000 to 10,000 kgf/mm² and a tensile strength of 105 to 175 kgf/mm² are included.

The difference between the claims and Peker is that Peker does not disclose a Young's modulus of 5,000 to 10,000 kgf/mm², a tensile strength of 105 to 175 kgf/mm², a relationship between the Young's modulus and tensile strength as defined by claim 30.

It would have been obvious to modify the face of Peker to have a Young's modulus and tensile strength as defined by the claims in order to have [a] face which has a sufficient flex for a specific golfer.

Appellants agree with the Examiner that Peker '642 fails to disclose a Young's modulus of 5,000 to 10,000 kgf/mm², a tensile strength of 105 to 175 kgf/mm², a relationship between the Young's modulus and tensile strength as defined by claim 30. However, Appellants disagree with the Examiner's assertion that it would be obvious to modify Peker '642 to arrive at the instant invention.

Appellants point out that Peker '642 provides no motivation or suggestion for modifying the composition of metals anywhere to arrive at the claimed invention.

Regarding motivation, the Examiner asserts that it would have been obvious to modify the face of Peker '642 to have a Young's modulus and tensile strength in order to have face, which has a sufficient flex for a specific golfer. The Examiner has not described that "specific golfer" and thus one of ordinary skill in the art would have no idea who that "specific golfer" would be. Moreover, the Examiner has also failed to indicate what is meant by "sufficient flex". Peker '642 fails to teach, describe, or even remotely suggest what is "sufficient flex for a specific golfer". Accordingly, Appellants submit that the requisite level of motivation necessary to make a *prima facie* case from the disclosure of Peker '642 is lacking.

Appellants would also like to submit that tensile strength is not an inherent property that is reliant only on the composition of the metal. As Appellants pointed in the response of May 7, 2003, the tensile strength is not only dependent on the composition of the metal but also on the method of annealing and methods of forging the metals in that composition. Appellants present as Appendix C two articles in Japanese filed with the Reply of May 7, 2003, with translations of the relevant parts, of how the method of making the metal plays a role in the tensile

strength. Accordingly, Appellants submit that one cannot simply arrive at the instant invention simply by providing the same composition as, for example, the embodiments in the instant invention. Thus, Appellants submit that the Examiner has also failed to meet the second requirement to make a proper *prima facie* rejection (i.e., expectation of success). In other words, one would not expect to arrive at the instant invention by the disclosure of Peker '642 without a disclosure of how that metal composition is going to be manufactured to arrive at a "sufficient flex for a specific golfer".

Moreover, Appellants submit the attached Appendix D wherein it is shown that the compositions disclosed in Peker '642 are not the same as the compositions as disclosed in the instant invention. Thus, not only does Peker '642 fail to disclose how one would manufacture the metal compositions of Peker '642 to provide "sufficient flex for a specific golfer" but Peker '642 also fails to disclose what compositions would provide "sufficient flex for a specific golfer".

Accordingly, Peker '642 cannot render obvious the instant invention because 1) Peker '642 does not provide the requisite motivation to arrive at the instant invention and 2) Peker '642 fails to disclose all the elements of the instantly claimed invention. Moreover, 3) one would not expect success from the disclosure of Peker '642 to arrive at the instant invention

because Peker '642 does not disclose how the composition of Peker '642 needs to be modified to arrive at the instant invention. Thus, a *prima facie* case of obviousness over the disclosure of Peker '642 has not been made. Reversal of the rejection of claims 21, 23-25 and 30 over Peker '642 is warranted and respectfully requested.

Argument 2

Claims 1-20, 22, 26-29, and 31-32 are patentable over Peker '642 in view of Kobayashi '742 (US Patent No. 5,611,742)

Appellants assert that the Examiner has failed to make out a *prima facie* case of obviousness with regard to the 35 USC §103(a) rejection of claims 1-20, 22, 26-29, and 31-32 over Peker '642 in view of Kobayashi '742. Three criteria must be met to make out a *prima facie* case of obviousness.

- 1) There must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings.
- 2) There must be a reasonable expectation of success.
- 3) The prior art reference (or references when combined) must teach or suggest all the claim limitations.

See MPEP §2142 and *In re Vaeck*, 20 USPQ2d 1438 (Fed. Cir. 1991). The Examiner has failed to meet any of these elements to make a

prima facie obviousness rejection. Claims 1 and 5 are the only independent claims that have been rejected in this rejection.

Claims 1 recites:

1. A golf club head comprising a hitting face for golf balls, said hitting face formed at least partially by a metallic material, and said metallic material satisfying the following relation:

$$y \geq 0.006x + 60$$

wherein

x is Young's modulus in units of kgf/mm^2 , and

y is tensile strength in units of kgf/mm^2 , and

wherein said metallic material has a young's modulus of 3,000 to 12,000 kgf/mm^2 , and a tensile strength of 105 to 175 kgf/mm^2 and

said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm.

Claim 5 recites:

5. A golf club head comprising a hitting face for golf balls, the surface of said hitting face being formed at least partially by a metallic material satisfying the following relationship:

$$z \geq (x/60) + 200$$

wherein x is Young's modulus in units of kgf/mm^2 , and z is Vickers hardness in units of HV, and

wherein said metallic material has a Young's modulus of 3,000 to 12,000 kgf/mm^2 and a Vickers hardness of 400 to 1,000 HV and said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm.

Appellants herein, present a table that shows some of the elements present in the above claims 1 and 5 and the comparable teachings in both Peker '642 and Kobayashi '742.

Claim 1	Peker '642	Kobayashi '742
a Young's modulus of 3,000 to 12,000	Peker '642 has no suggestion or	Kobayashi '742 has no suggestion or

kgf/mm ²	disclosure of Young's modulus.	disclosure of Young's modulus.
A tensile strength of 105 to 175 kgf/mm ²	The only disclosure in Peker '642 of a tensile strength is 194 kgf/mm ² .	Kobayashi '742 discloses a tensile strength of 65 kgf/mm ² .
Claim 5	Peker '642	Kobayashi '742
a Young's modulus of 3,000 to 12,000 kgf/mm ²	Peker '642 has no suggestion or disclosure of Young's modulus.	Kobayashi '742 has no suggestion or disclosure of Young's modulus.
A Vickers hardness of 400 to 1,000 HV	Peker '642 has no suggestion or disclosure of Vickers hardness.	Kobayashi '742 has no suggestion or disclosure of Vickers hardness.

Regarding claim 1, Appellants assert that neither Peker '642 nor Kobayashi '742 suggest or disclose the claimed features of the instant invention. In particular, neither Peker '642 nor Kobayashi '742 suggest or disclose anything regarding a Young's modulus. Moreover, neither Peker '642 nor Kobayashi '742 disclose or suggest the claimed range of tensile strength. The only disclosure in Peker '642 of a tensile strength is a tensile strength of 194 kgf/mm² (see column 4, lines 28-29 in Peker '642). Kobayashi '742, in table 1 in columns 3 and 4 discloses tensile strengths that are from 53 kgf/mm² to 65 kgf/mm² (to convert from N/mm² to kgf/mm², one must divide by 10). Appellants point out that all of these tensile strengths in Kobayashi '742 fall outside of the claimed range in claim 1. Thus, Appellants submit that the Examiner has failed to make out

a *prima facie* case with respect to the rejection over Peker '642 in view of Kobayashi '742 because all of the elements of the instantly claimed invention have not been disclosed or remotely suggested by the combination of the disclosures of Peker '642 and Kobayashi '742.

Moreover, Appellants assert that neither Peker '642 nor Kobayashi '742 provide the requisite motivation in either of their disclosures to arrive at the instant invention. Peker '642 and Kobayashi '742, as pointed out above, disclose tensile strengths that fall outside of the scope of the range of tensile strengths claimed in claim 1. Neither Peker '642 nor Kobayashi '742 remotely suggest modifying the tensile strengths to arrive at the instant invention. Moreover, neither Peker '642 nor Kobayashi '742 disclose or remotely suggest having a Young's modulus that falls within the scope of the claimed range in claim 1 of the instant invention.

The combination of Peker '642 and Kobayashi '742 also does not provide any expectation of success at arriving at the instant claim 1. Because neither Peker '642 nor Kobayashi '742 disclose or suggest the tensile strength claimed in claim 1, one would not expect to attain the instant invention without a disclosure of modifying said tensile strength. As pointed out above, tensile strength is not an inherent property that is reliant only on the composition of the metal. As Appellants

pointed in the response of May 7, 2003, the tensile strength is not only dependent on the composition of the metal but also on the method of annealing and methods of forging the metals in that composition. Appellants present as Appendix C two articles in Japanese filed with the Reply of May 7, 2003, with translations of the relevant parts, of how the method of making the metal plays a role in the tensile strength and Vickers hardness. Accordingly, Appellants submit that one cannot simply arrive at the instant invention simply by providing the same composition as, for example, the embodiments in the instant invention. Thus, Appellants submit that the Examiner has also failed to meet the second requirement to make a proper *prima facie* rejection (i.e., expectation of success).

For the above reasons, claim 1 cannot be rendered obvious by the disclosures of Peker '642 in view of Kobayashi '742 because the two references when combined together fail to render the instant invention *prima facie* obvious. Reversal of the rejection of claim 1 over Peker '642 in view of Kobayashi '742 is warranted and respectfully requested.

Regarding claim 5, Appellants submit that the Examiner has also failed to show why the instant invention is rendered *prima facie* obvious by Peker '642 in view of Kobayashi '742. Neither Peker '642 nor Kobayashi '742 disclose or suggest a Young's modulus of 3,000 to 12,000 kgf/mm² or a Vickers hardness of 400

to 1,000 HV. Thus, the elements of the instant invention have not been met by the disclosure of Peker '642 in view of Kobayashi '742. Moreover, Appellants assert that Vickers hardness is a property that is not inherent based simply on the composition of the metal. A Vickers hardness is calculated by dividing a load placed on a metal by the surface area of an impression that that load generates. The surface area of the impression is dependent on the structure of the alloy composition, which in turn is dependent on how the alloy is manufactured. Thus, the Vickers hardness is not simply dependent on the composition of the metal. Please see Appendix C which is two articles in Japanese filed with the Reply of May 7, 2003, with translations of the relevant parts, of how the method of making the metal plays a role in the tensile strength and Vickers hardness. For these reasons, the Vickers hardness is not inherent to a given metal composition. Thus, absent a disclosure of the Vicker's hardness one cannot arrive at the instant invention by the combination of Peker '642 and Kobayashi '742.

The combination of Peker '642 and Kobayashi '742 cannot render obvious the instant invention because all of the elements of the instant invention are not disclosed or suggested, one would not expect success at arriving at the instant invention from their disclosures, and there is no motivation or suggestion

to modify either of Peker '642 or Kobayashi '742 (or their combination) to arrive at the instant invention. Reversal of the rejection with respect to claim 5 over Peker '642 in view of Kobayashi '742 is warranted and respectfully requested.

Argument 3

Claims 42-43 are patentable over Peker '642 in view of Kobayashi '742 and further in view of Anderson '663 (US Patent No. 5,261,663)

Appellants assert that the Examiner has failed to make out a *prima facie* case of obviousness with regard to the 35 USC §103(a) rejection over Anderson '663. Three criteria must be met to make out a *prima facie* case of obviousness.

1) There must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings.

2) There must be a reasonable expectation of success.

3) The prior art reference (or references when combined) must teach or suggest all the claim limitations.

See MPEP §2142 and *In re Vaeck*, 20 USPQ2d 1438 (Fed. Cir. 1991).

The Examiner has failed to meet any of the elements necessary to make a *prima facie* obviousness rejection.

Claims 42 and 43, respectively, recite:

42. The golf club head of claim 1 wherein the head comprises a head body and a face plate made of said metallic material and the head body is provided with a face mounting part for attaching the face plate comprising a periphery of a hitting face, and the face mounting part is provided with a step down zone.

43. The golf club head of claim 5 wherein the head comprises a head body and a face plate made of said metallic material and the head body is provided with a face mounting part for attaching the face plate comprising a periphery of a hitting face, and the face mounting part is provided with a step down zone.

Appellants point out that claims 42 and 43 are dependent from claims 1 and 5, respectively. Claim 42 cannot be rendered obvious for the same reasons why Peker '642 and Kobayashi '742 cannot render obvious claim 1. In the below table, Appellants point out some of the elements present in claim 1 and where the corresponding teachings are present or not present in Peker '642, Kobayashi '742 and Anderson '663.

Claim 1	Peker '642	Kobayashi '742	Anderson '663
a Young's modulus of 3,000 to 12,000 kgf/mm ²	Peker '642 has no suggestion or disclosure of Young's modulus.	Kobayashi '742 has no suggestion or disclosure of Young's modulus.	Anderson '663 has no suggestion or disclosure of Young's modulus.
A tensile strength of 105 to 175 kgf/mm ²	The only disclosure in Peker '642 of a tensile strength is 194 kgf/mm ² .	Kobayashi '742 discloses a tensile strength of 65 kgf/mm ² .	Anderson '663 has no suggestion or disclosure of tensile strength.

In the above table, it should be apparent to one of ordinary skill in the art that Anderson '663 fails to make up the deficiencies present in Peker '642 and Kobayashi '742. None of Anderson '663, Peker '642 or Kobayashi '742 discloses or suggests the Young's modulus or the tensile strength that are claimed in claim 1. Anderson '663, Peker '642 and Kobayashi '742 fail to say anything about the Young's modulus of the metal composition for the golf club head. Peker '642 and Kobayashi '742 disclose tensile strengths of 194 kgf/mm² and 65 kgf/mm² (at column 4, lines 28-29 in Peker '642 and in Table 1 in columns 3 and 4 in Kobayashi '742, respectively). Both of these disclosed tensile strengths fall outside of the claimed tensile strength range of 105 to 175 kgf/mm² in claim 1. Because Anderson '663 says nothing of tensile strength, Anderson '663 fails to make up for the deficiencies of Peker '642 and Kobayashi '742. Thus, because all of the elements of claim 42 is not disclosed, a *prima facie* case of obviousness has not been made by the combination of Anderson '663, Peker '642 and Kobayashi '742. Accordingly, the rejection is inapposite. Reversal of the Examiner in regards to the rejection of claim 42 with respect to the combination of Anderson '663, Peker '642 and Kobayashi '742 is warranted and respectfully requested.

Regarding claim 43, Appellants submit that combination of Anderson '663, Peker '642 and Kobayashi '742 also cannot render this claim *prima facie* obvious. None of Anderson '663, Peker '642

or Kobayashi '742 discloses or suggests the elements of claim 5, from which claim 43 is dependent. The below table illustrates this.

Claim 5	Peker '642	Kobayashi '742	Anderson '663
a Young's modulus of 3,000 to 12,000 kgf/mm ²	Peker '642 has no suggestion or disclosure of Young's modulus.	Kobayashi '742 has no suggestion or disclosure of Young's modulus.	Anderson '663 has no suggestion or disclosure of Young's modulus
a Vickers hardness of 400 to 1,000 HV	Peker '642 has no suggestion or disclosure of Vickers hardness.	Kobayashi '742 has no suggestion or disclosure of Vickers hardness.	Anderson '663 has no suggestion or disclosure of Vickers hardness.

As can be seen by the above table, the combination of Anderson '663, Peker '642 and Kobayashi '742 fails to disclose or suggest the elements present in claim 5. Thus, the combination of Anderson '663, Peker '642 and Kobayashi '742 cannot render obvious claim 43, which depends from claim 5, because all of the elements of claim 5 are not disclosed or even remotely suggested.

Moreover, as was explained above, Vickers hardness is not an inherent property that relies only on the composition. The Vickers hardness of a composition metal is also dependent on the process by which the final product is made. Because the Vickers hardness is partially process dependent and because Anderson '663, Peker '642 and Kobayashi '742 say nothing about Vickers hardness,

Anderson '663, Peker '642 and Kobayashi '742 cannot render obvious claim 43. Reversal of the rejection with respect to claim 43 in view of the combination of Anderson '663, Peker '642 and Kobayashi '742 is warranted and respectfully requested.

Argument 4

Claims 1, 5, and 45-46 are patentable over Peker '642 in view of Kobayashi '501 (US Patent No. 5,601,501) and further in view of Sieleman '005 (US Patent No. 5,792,005)

Appellants assert that the Examiner has failed to make out a *prima facie* case of obviousness with regard to the 35 USC §103(a) rejection of claims 1, 5 and 45-46 over Peker '642 in view of Kobayashi '501 and further in view of Sieleman '005. Three criteria must be met to make out a *prima facie* case of obviousness.

- 1) There must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings.
- 2) There must be a reasonable expectation of success.
- 3) The prior art reference (or references when combined) must teach or suggest all the claim limitations.

See MPEP §2142 and *In re Vaeck*, 20 USPQ2d 1438 (Fed. Cir. 1991). The Examiner has failed to meet any of these elements to make a

prima facie obviousness rejection. Claims 1 and 5 are the only independent claims that have been rejected in this rejection.

Claims 1 recites:

1. A golf club head comprising a hitting face for golf balls, said hitting face formed at least partially by a metallic material, and said metallic material satisfying the following relation:

$$y \geq 0.006x + 60$$

wherein

x is Young's modulus in units of kgf/mm^2 , and

y is tensile strength in units of kgf/mm^2 , and

wherein said metallic material has a young's modulus of 3,000 to 12,000 kgf/mm^2 , and a tensile strength of 105 to 175 kgf/mm^2 and

said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm.

Claim 5 recites:

5. A golf club head comprising a hitting face for golf balls, the surface of said hitting face being formed at least partially by a metallic material satisfying the following relationship:

$$z \geq (x/60) + 200$$

wherein x is Young's modulus in units of kgf/mm^2 , and z is Vickers hardness in units of HV, and

wherein said metallic material has a Young's modulus of 3,000 to 12,000 kgf/mm^2 and a Vickers hardness of 400 to 1,000 HV and said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm.

The below table illustrates some of the elements that are present in claims 1 and 5 and where the corresponding elements are found or not found in Peker '642, Kobayashi '501 and Sieleman '005.

Claim 1	Peker '642	Kobayashi '501	Sieleman '005
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a Young's modulus of 3,000 to 12,000 kgf/mm ²	Peker '642 has no suggestion or disclosure of Young's modulus.	Kobayashi '501 has no suggestion or disclosure of Young's modulus.	Sieleman '005 has no suggestion or disclosure of Young's modulus.
a tensile strength of 105 to 175 kgf/mm ²	The only disclosure in Peker '642 of a tensile strength is 194 kgf/mm ² .	The disclosure in Kobayashi '501 of tensile strengths is from 53 to 65 kgf/mm ² .	Sieleman '005 has no suggestion or disclosure of tensile strength.
Claim 5	Peker '642	Kobayashi '501	Sieleman '005
a Young's modulus of 3,000 to 12,000 kgf/mm ²	Peker '642 has no suggestion or disclosure of Young's modulus.	Kobayashi '501 has no suggestion or disclosure of Young's modulus.	Sieleman '005 has no suggestion or disclosure of Young's modulus.
a Vickers hardness of 400 to 1,000 HV	Peker '642 has no suggestion or disclosure of Vickers hardness.	Kobayashi '501 has no suggestion or disclosure of Vickers hardness.	Sieleman '005 has no suggestion or disclosure of Vickers hardness.

From the above table, it should be apparent to one of ordinary skill in the art that the combination of Peker '642, Kobayashi '501 and Sieleman '005 fail to disclose all of the elements of claims 1 and 5. Accordingly, Peker '642, Kobayashi '501 and Sieleman '005 cannot render obvious the instant invention because they fail to render claims 1, 5, and 45-46 *prima facie* obvious.

In particular, none of Peker '642, Kobayashi '501 and Sieleman '005 disclose or suggest the tensile strength disclosed

in claim 1. Peker '642 (194 kgf/mm², see column 4, lines 28-29) and Kobayashi '501 (53 to 65 kgf/mm², see Table 1 in columns 3 and 4) disclose a tensile strength range that is outside the scope of the range claimed in claim 1 (i.e., 105 to 175 kgf/mm²). Sieleman '005 fails to disclose or even remotely suggest any tensile strength. Thus, Sieleman '005 does not cure the deficiencies present in Peker '642 and Kobayashi '501 necessary to arrive at the instant invention. Thus, Appellants assert that a *prima facie* case of obvious over the combination of Peker '642, Kobayashi '501 and Sieleman '005 has not been made because all the elements of claim 1 have not been disclosed.

Because all of the elements in claim 1 have not been disclosed, one of ordinary skill in the art would have no expectation of success in arriving at the instant invention. Moreover, there is no suggestion or motivation in any of the references for modifying them to arrive at the instant invention. Thus, Appellants assert that a *prima facie* case of obvious over the combination of Peker '642, Kobayashi '501 and Sieleman '005 has not been made for either claim 1, or claim 45, which is dependent from claim 1.

Regarding claim 5 and 46, Peker '642, Kobayashi '501 and Sieleman '005 fail to disclose or remotely suggest either of a Young's modulus or a Vickers hardness as is present in the claim. Moreover, as was explained above, Vickers hardness is

not an inherent property that is only reliant on the composition of the metal. The Vickers hardness is also dependent on the process of making that metal alloy. Please see Appendix C, which is two articles in Japanese filed with the Reply of May 7, 2003, with translations of the relevant parts, of how the method of making the metal plays a role in the Vickers hardness. Thus, because Vickers hardness is not an inherent property of the metal composition, a *prima facie* case of obviousness has not been established relative to claim 5. Thus, claim 46, which is dependent from claim 5 also cannot be rendered *prima facie* obvious over the disclosures of Peker '642, Kobayashi '501 and Sieleman '005.

Reversal of the Examiner's rejection of claims 1, 5, and 45-46 over Peker '642, Kobayashi '501 and Sieleman '005 is warranted and respectfully requested.

IX. Conclusion

For the reasons advanced above, it is respectfully submitted that all claims in this application are allowable. Thus, favorable reconsideration and reversal of the Examiner's rejection of claims 1-38 and 42-46 under 35 U.S.C. § 103, for allegedly being unpatentable over the cited references, by the Honorable Board of Patent Appeals and Interferences, is respectfully solicited.

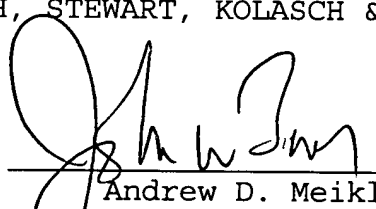
The required Appeal Brief fee in the amount of \$330.00 is attached hereto. An extension of two (2) month(s) was previously requested and paid for on October 30, 2003 in the instant application. Thus, no extension fee is required.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

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Attachment: APPENDIX A: Claims as appealed

APPENDIX B: Tables 1 and 2 filed with Reply of May 7, 2003

APPENDIX C: Japanese articles and translation of relevant portions filed with Reply of May 7, 2003

APPENDIX A

1. (previously presented) A golf club head comprising a hitting face for golf balls, said hitting face formed at least partially by a metallic material, and said metallic material satisfying the following relation:

$$y \geq 0.006x + 60$$

wherein

x is Young's modulus in units of kgf/mm², and

y is tensile strength in units of kgf/mm², and

wherein said metallic material has a young's modulus of 3,000 to 12,000 kgf/mm², and a tensile strength of 105 to 175 kgf/mm² and said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm.

2. (previously presented) A golf club head according to claim 1, wherein said metallic material is an amorphous metal.

3. (previously presented) A golf club head according to claim 1, wherein said metallic material is an amorphous alloy of a zirconium base.

4. (previously presented) A golf club head according to claim 1, wherein said metallic material is an amorphous alloy comprising

the elements Zr, Al, Cu, Ni, and Hf or an amorphous alloy comprising the elements Zr, Al, Cu, and Ni.

5. (previously presented) A golf club head comprising a hitting face for golf balls, the surface of said hitting face being formed at least partially by a metallic material satisfying the following relationship:

$$z \geq (x/60) + 200$$

wherein x is Young's modulus in units of kgf/mm^2 , and z is Vickers hardness in units of HV, and

wherein said metallic material has a Young's modulus of 3,000 to 12,000 kgf/mm^2 and a Vickers hardness of 400 to 1,000 HV and said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm.

6. (previously presented) A golf club head according to claim 5, wherein said metallic material is an amorphous metal.

7. (previously presented) A golf club head according to claim 5, wherein said metallic material is an amorphous alloy of a zirconium base.

8. (previously presented) A golf club head according to claim 1, wherein said metallic material is an amorphous alloy comprising the elements Zr, Al, Cu, Ni, and Hf or an amorphous alloy comprising the elements Zr, Al, Cu, and Ni.

9. (previously presented) A golf ball club head according to claim 1, wherein said metallic material has a Young's modulus of 5,000 to 12,000 kgf/mm² and a tensile strength of 105 to 400 kgf/mm².

10. (previously presented) A golf ball club head according to claim 1, wherein said metallic material has a Young's modulus of 5,000 to 12,000 kgf/mm² and a tensile strength of 130 to 400 kgf/mm².

11. (previously presented) A golf ball club head according to claim 5, wherein said metallic material has a Young's modulus of 5,000 to 12,000 kgf/mm² and a Vickers hardness of 400 to 1,000 HV.

12. (previously presented) A golf ball club head according to claim 5, wherein said metallic material has a Young's modulus of 5,000 to 12,000 kgf/mm² and a Vickers hardness of 400 to 1,000 HV.

13. (previously presented) A golf ball club head according to claim 5, wherein said metallic material has a tensile strength of 80 to 400 kgf/mm².

14. (previously presented) A golf ball club according to claim 1, wherein the metallic metal is an amorphous metal expressed by the formula:

M_aX_b , wherein M represents two or more elements selected from the group consisting of Zr, V, Cr, Mn, Fe, Co, Ni, Cu, Ti, Mo, W, Ca, Li, Mg, Si, Al, Pd and Be; X is an element selected from the group consisting of Y, La, Ce, Sm, Md, Hf, Nb and Ta; and a and b represent atomic percentages in the ranges of $65 \leq a \leq 100$ and $0 \leq b \leq 35$, respectively.

15. (previously presented) A golf ball club according to claim 5, wherein the metallic metal is an amorphous metal expressed by the formula:

M_aX_b , wherein M represents two or more elements selected from the group consisting of Zr, V, Cr, Mn, Fe, Co, Ni, Cu, Ti, Mo, W,

Ca, Li, Mg, Si, Al, Pd and Be; X is an element selected from the group consisting of Y, La, Ce, Sm, Md, Hf, Nb and Ta; and a and b represent atomic percentages in the ranges of $65 \leq a \leq 100$ and $0 \leq b \leq 35$, respectively.

16. (previously presented) A golf ball head according to claim 1 wherein the metallic material is an amorphous metal of the formula: $Zr_cM_dX_e$, wherein Zr is Zirconium; M is an element selected from the group consisting of V, Cr, Mn, Fe, Co, Ni, Cu, Ti, Mo, W, Ca, Li, Mg, Si, Al, Pd and Be; X is an element selected from the group consisting of Y, La, Ce, Sm, Md, Hf, Nb and Ta; and c, d and e represent atomic percentages within the ranges of $20 \leq c \leq 80$, $20 \leq d \leq 80$, and $0 \leq e \leq 35$, respectively.

17. (previously presented) A golf ball head according to claim 5 wherein the metallic material is an amorphous metal of the formula: $Zr_cM_dX_e$, wherein Zr is Zirconium; M is an element selected from the group consisting of V, Cr, Mn, Fe, Co, Ni, Cu, Ti, Mo, W, Ca, Li, Mg, Si, Al, Pd and Be; X is an element selected from the group consisting of Y, La, Ce, Sm, Md, Hf, Nb and Ta; and c, d and e represent atomic percentages within the ranges of $20 \leq c \leq 80$, $20 \leq d \leq 80$, and $0 \leq e \leq 35$, respectively.

18. (previously presented) A golf ball head according to claim 1 wherein the metallic material is an amorphous metal of the formula: $Zr_cM_dX_e$, wherein Zr is Zirconium; M is an element selected from the group consisting of V, Cr, Mn, Fe, Co, Ni, Cu, Ti, Mo, W, Ca, Li, Mg, Si, Al, Pd and Be; X is an element selected from the group consisting of Y, La, Ce, Sm, Nd, Hf, Nb and Ta; and c, d and e represent atomic percentages within the ranges of $50 \leq c \leq 75$, $25 \leq d \leq 50$, and $0 \leq e \leq 1$, respectively.

19. (previously presented) A golf ball head according to claim 5 wherein the metallic material is an amorphous metal of the formula: $Zr_cM_dX_e$, wherein Zr is Zirconium; M is an element selected from the group consisting of V, Cr, Mn, Fe, Co, Ni, Cu, Ti, Mo, W, Ca, Li, Mg, Si, Al, Pd and Be; X is an element selected from the group consisting of Y, La, Ce, Sm, Nd, Hf, Nb and Ta; and c, d and e represent atomic percentages within the ranges of $50 \leq c \leq 75$, $25 \leq d \leq 50$, and $0 \leq e \leq 1$, respectively.

20. (previously presented) A golf club head according to claim 5, wherein a thickness of said metallic material is 1 to 3 mm.

21. (previously presented) A golf club head comprising a hitting face for golf balls, said hitting face formed at least

partially by a metallic material, and said metallic material satisfying the following relationship:

$$y \geq 0.006x + 60$$

wherein x is Young's modulus in units of kgf/mm^2 , and y is tensile strength in units kgf/mm^2 , and

wherein said metallic material has a Young's modulus of 5,000 to 16,000 kgf/mm^2 and a tensile strength of 105 to 175 kgf/mm^2 .

22. (previously presented) A golf club head according to claim 21, wherein a thickness of said metallic material is 1 to 3 mm.

23. (previously presented) A golf club head according to claim 21, wherein said metallic material is an amorphous metal.

24. (previously presented) A golf club head according to claim 21, wherein said metallic material is an amorphous alloy of a zirconium base.

25. (previously presented) A golf club head according to claim 21, wherein said metallic material is an amorphous alloy comprising the elements Zr, Al, Cu, Ni, and Hf or an amorphous alloy comprising the elements Zr, Al, Cu and Ni.

26. (previously presented) A golf club head according to claim 7, wherein said metallic material satisfies the following relation:

$y > 0.006x + 63$ wherein y is tensile strength in units of kgf/mm^2 .

27. (previously presented) A golf club head according to claim 1, wherein the back of said hitting portion is not supported by a support member.

28. (previously presented) A golf club head according to claim 5, wherein said metallic material has a young's modulus of 3,000 to 10,000 kgf/mm^2 .

29. (previously presented) A golf club head according to claim 5, wherein the back of said hitting portion is not supported by a support member.

30. (previously presented) A golf club head according to claim 21, wherein said metallic material satisfies the following relation:

$y > 0.006x + 63$.

31. (previously presented) A golf club head according to claim 21, wherein said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm.

32. (previously presented) A golf club head according to claim 21, wherein said hitting face has at least partially a hitting portion which consists of said metallic material with a thickness of 1 to 3 mm and the back of said hitting portion is not supported by a support member.

33. (previously presented) The golf club head of claim 1 wherein the head is wood.

34. (previously presented) The golf club head of claim 5 wherein the head is wood.

35. (previously presented) The golf club head of claim 21 wherein the head is wood.

36. (previously presented) The golf club head of claim 1 wherein the head is iron.

37. (previously presented) The golf club head of claim 5 wherein the head is iron.

38. (previously presented) The golf club head of claim 21 wherein the head is iron.

39. (withdrawn - previously presented) The golf club head of claim 1 wherein the hitting portion has uniform thickness.

40. (withdrawn - previously presented) The golf club head of claim 5 wherein the hitting portion has uniform thickness.

41. (withdrawn - previously presented) The golf club head of claim 21 wherein the hitting portion has uniform thickness.

42. (previously presented) The golf club head of claim 1 wherein the head comprises a head body and a face plate made of said metallic material and the head body is provided with a face mounting part for attaching the face plate comprising a periphery of a hitting face, and the face mounting part is provided with a step down zone.

43. (previously presented) The golf club head of claim 5 wherein the head comprises a head body and a face plate made of said

metallic material and the head body is provided with a face mounting part for attaching the face plate comprising a periphery of a hitting face, and the face mounting part is provided with a step down zone.

44. (previously presented) The golf club head of claim 21 wherein the head comprises a head body and a face plate made of said metallic material and the head body is provided with a face mounting part for attaching the face plate comprising a periphery of a hitting face, and the face mounting part is provided with a step down zone. ✓

45. (previously presented) The golf club head of claim 1 wherein the head comprises a head body and a face plate made of said metallic material wherein the face plate is constructed with a thicker central part with a periphery part whose thickness reduces gradually outward. ✓

46. (previously presented) The golf club head of claim 5 wherein the head comprises a head body and a face plate made of said metallic material wherein the face plate is constructed with a thicker central part with a periphery part whose thickness reduces gradually outward. ✓

47. (canceled)

48. (withdrawn - previously presented) The golf club head of claim 1 wherein the head comprises a head body and a face plate made of said metallic material wherein the face plate is constructed with a thinner central part with a periphery part whose thickness increases gradually outward.

49. (withdrawn - previously presented) The golf club head of claim 5 wherein the head comprises a head body and a face plate made of said metallic material wherein the face plate is constructed with a thinner central part with a periphery part whose thickness increases gradually outward.

50. (withdrawn - previously presented) The golf club head of claim 21 wherein the head comprises a head body and a face plate made of said metallic material wherein the face plate is constructed with a thinner central part with a periphery part whose thickness increases gradually outward.

Table 1

	Present Invention	Peker	Aizawa
claim1	Young's Modulus: $x \text{ kgf/mm}^2$ Tensile strength: $y \text{ kgf/mm}^2$ $y \geq 0.006x + 60$	No	No
	$x : 3000 \text{ to } 12000 \text{ kgf/mm}^2$	No	$\text{No} : 30000 \text{ kgf/mm}^2$
	$y : 105 \text{ to } 175 \text{ kgf/mm}^2$ Hitting portion which consists of metallic material has a thickness of 1 to 3 mm.	$\text{No} : 194 \text{ kgf/mm}^2$	No
claim5	Young's Modulus: $x \text{ kgf/mm}^2$ Vickers Hardness: $z \text{ HV}$ $z \geq (x/60) + 200$	No	The each face plates shown in Fig.2 and 4 is surpoted by the surport wall. The face plate shown in Fig.5 has a thickness of more than 5mm. No
	$x : 3000 \text{ to } 12000 \text{ kgf/mm}^2$	No	$\text{No} : 30000 \text{ kgf/mm}^2$
	$y : 400 \text{ to } 1000 \text{ HV}$ Hitting portion which consists of metallic material has a thickness of 1 to 3 mm.	No	No
claim21	Young's Modulus: $x \text{ kgf/mm}^2$ Tensile strength: $y \text{ kgf/mm}^2$ $y \geq 0.006x + 60$	No	The each face plates shown in Fig.2 and 4 is surpoted by the surport wall. The face plate shown in Fig.5 has a thickness of more than 5mm. No
	$x : 5000 \text{ to } 16000 \text{ kgf/mm}^2$	No	$\times : 30000 \text{ kgf/mm}^2$
	$y : 105 \text{ to } 175 \text{ kgf/mm}^2$	$\text{No} : 194 \text{ kgf/mm}^2$	No

Table 2

Alloys composition of Peker

composition	Zr	Ti	Be	Ni	Cu	Fe	Co	Mn	Hf	Al	Mo	Cr
type-a	41.2	13.8	22.5	10	12.5							
type-b	46.75	8.25	27.5	10	7.5							
type-c	25~85				5~70				5~35			
type-d	60			25							15	

Alloys composition of the embodiment of this present invention

composition	Zr	Ti	Be	Ni	Cu	Fe	Co	Mn	Hf	Al	Mo	Cr
ex.1	54				5	30				1	10	
ex.2,11	64				10	15				1	10	
ex.3,4,6,9,12	55				5	30					10	
ex.5	50				10	20					10	
ex.7,10	55				10	25					10	
ex.8	54				10	15				1	10	

APPENDIX C

Publication 1

Akita University mining department research report No. 14
issued in October, 1993

The translation of the A section

Fig.12 shows change of the tensile strength (σ) and hardness (Hv) by the annealing temperature of $Al_{9.8}Ce_1Co_1$, $Al_{9.6}Ce_2Co_3$ and $Al_{9.7}Ce_2Co_1$ alloy. Although the influence of annealing of is small in the case of low dissolution concentration like the $Al_{98}Ce_1Co_1$ alloy, if the amount of Ce or Co increase, the temperature to which σ falls will shift to a high temperature side, σ of $Al_{96}Ce_1Co_3$ and $Al_{97}Ce_2Co_1$ alloy rapidly decreases from the 573K neighborhood, and σ becomes an almost fixed value and has become the substantially same value as $Al_{98}Ce_1Co_1$ alloy from more than 673K.

The translation of the B section

Next, hardness is described. In the case of $Al_{98}Ce_1Co_1$ alloy with few amounts of each Ce and Co, Hv is slowly decreasing in connection with rising of the annealing temperature, and both $Al_{96}Ce_2Co_3$ and $Al_{97}Ce_2Co_1$ alloys are decreasing from the 473K neighborhood. This is influenced by separating of a compound and it is considered to receive influence also by removing lattice strain gradually with reduction of the various defects in a rapid cooling state. Since there is little these influence in the case of the fcc-Al single phase alloy of low dissolution concentration, it is thought that the degree of change of Hv is small.

論文

液体急冷 Al-Ce-Co 合金の組織と機械的性質*

渡部 充**・高橋 太***・永田 明彦**
木村 久道****・井上 明久****・増本 健****

Structure and Mechanical Properties of
Rapidly Solidified Al-Ce-Co Alloys.

Mitsuru WATANABE**, Futoshi TAKAHASHI***, Akihiko NAGATA**,
Hisamichi KIMURA****, Akihisa INOUE**** and Tsuyoshi MASUMOTO****

Abstract

Experiments on structure and mechanical properties of rapidly solidified and annealed Al-Ce-Co alloys were investigated. The results obtained were as follows.

- (1) With increasing solute content, the rapidly solidified structure changes in the order of fcc solid solution (Al), fcc Al plus amorphous and amorphous phases. The fcc Al phase was formed in solute concentration (Ce+Co) range below about 5 at%.
- (2) The rapidly solidified alloys were almost ductile.
- (3) Crystallization or decomposition temperature (T_x) of the rapidly solidified alloys was almost high in the fcc Al phase of lower Co content and higher Ce content, but low in the amorphous phase.
- (4) $Al_{97}Ce_2Co_1$ alloy of the fcc Al phase and $Al_{96}Ce_4Co_{10}$ alloy of the amorphous phase showed high tensile strength, and the former showed high hardness value.
- (5) Alloys of the fcc Al phase were ductile after annealing for 3.6 ks at 373~773 K. On the other hand, alloys of the amorphous phase were brittle after annealing for 3.6 ks at 473~773 K, except $Al_{94}Ce_5Co_1$ alloy.
- (6) Tensile strength and hardness of alloys of the fcc Al phase decreased by annealing treatment.

1. 緒 言

最近, 液体急冷による高比強度 Al 基材料の開発に関する研究がなされている。急冷効果と固溶強化に大きな影響を及ぼす元素として Ce が考えられ, Al-Ce-M (M = 遷移金属) 系 3 元合金における相領域, 熱的安定性および機械的性質などについて報告がなされている。Ce は原子半径が Al よりかなり大きく, しかも従来の溶解鑄造法では Al に固溶しない。したがって, 急冷効果が大きいことにより, 液体急冷凝固法によって Al に強制的に固溶することがで

Cerium
58
(57-58)

1993 年 6 月 25 日受理

* 日本材料学会にて一部講演 (1993 年 5 月, 仙台)

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sity.

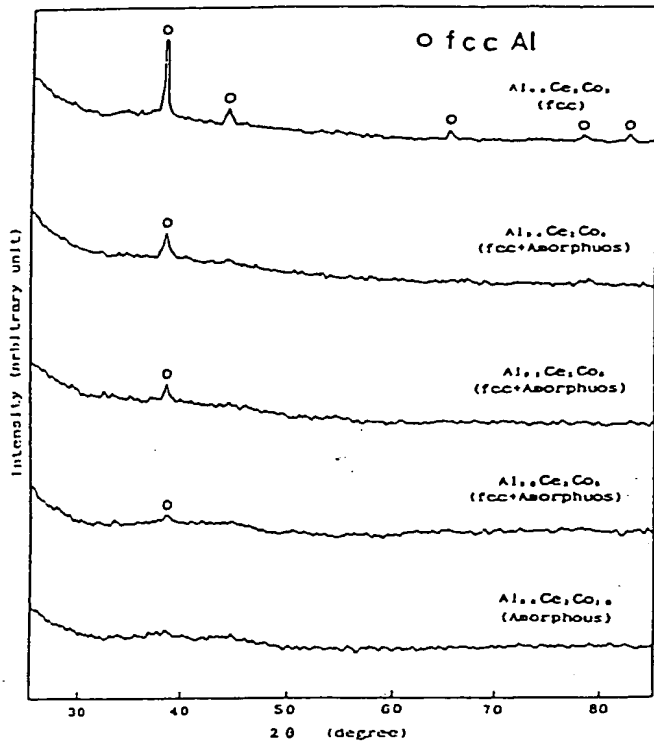


Fig. 1 X-ray diffraction patterns of rapidly solidified $\text{Al}_{98-x}\text{Ce}_2\text{Co}_x$ ($x = 2, 4, 6, 8, 10$) alloys.

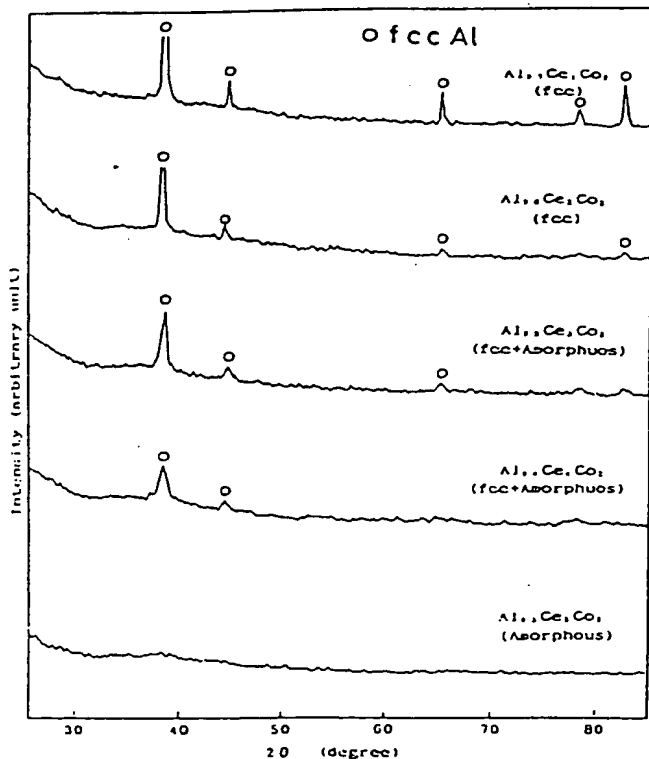


Fig. 2 X-ray diffraction patterns of rapidly solidified $\text{Al}_{98-x}\text{Ce}_x\text{Co}_2$ ($x = 1, 2, 3, 4, 5$) alloys.

図を作成した。この点については後述する。

3.2 急冷材の電子顕微鏡観察

Fig. 3(a)~(c)に液体急冷した $\text{Al}_{96}\text{Ce}_2\text{Co}_2$, $\text{Al}_{95}\text{Ce}_3\text{Co}_2$ および $\text{Al}_{95}\text{Ce}_4\text{Co}_1$ 合金の透過電子顕微鏡写真を示す。左が明視野像、右は制限視野回折パターンである。X線回折パターンで示したように、 $\text{Al}_{96}\text{Ce}_2\text{Co}_2$ 合金は電子回折パターンでも明瞭な fcc Al 相のスポットが認められ、fcc Al 単相であることが確認できる。 $\text{Al}_{95}\text{Ce}_3\text{Co}_2$ 合金の場合には fcc Al 相の回折スポットがうすれてきている。また、 $\text{Al}_{95}\text{Ce}_4\text{Co}_1$ 合金ではさらに fcc Al 相の回折スポットが少なくなり、X線回折パターンと併せて考えれば、一部スポットがストリークしてきており、アモルファス相が次第に生成されてきていることを示唆しているものと考えられる。

3.3 示差走査熱量測定

Fig. 4 に液体急冷材の DSC 曲線の代表的な例として、 $\text{Al}_{99-x}\text{Ce}_1\text{Co}_x$ ($x = 1, 2, 3, 4, 5, 8, 10$) 合金の DSC 曲線を示す。図より 1 at%Ce の場合 Co 量が多くなるに従って、結晶化または分解温度 (T_x) が低温側に移行する傾向を示している。すなわち、アモルファス相の増加に伴って T_x が低くなり、結晶化によ

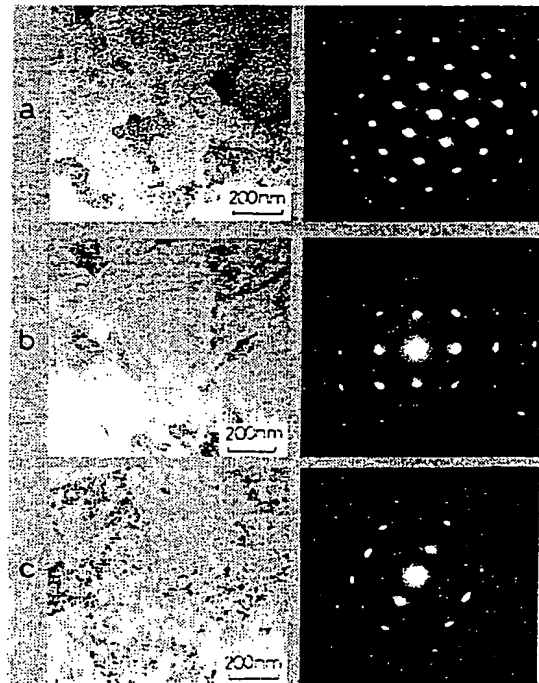


Fig. 3 Bright-field electron micrographs (left) and selected area diffraction patterns (right) of rapidly solidified $\text{Al}_{96}\text{Ce}_2\text{Co}_2$ (a), $\text{Al}_{95}\text{Ce}_3\text{Co}_2$ (b) and $\text{Al}_{95}\text{Ce}_4\text{Co}_1$ (c) alloys.

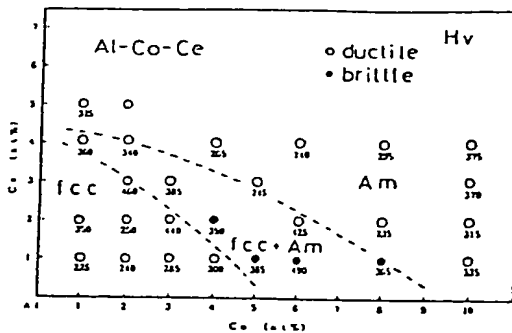


Fig. 7 Compositional dependences of Vickers hardness (H_v) for rapidly solidified Al-Ce-Co alloys.

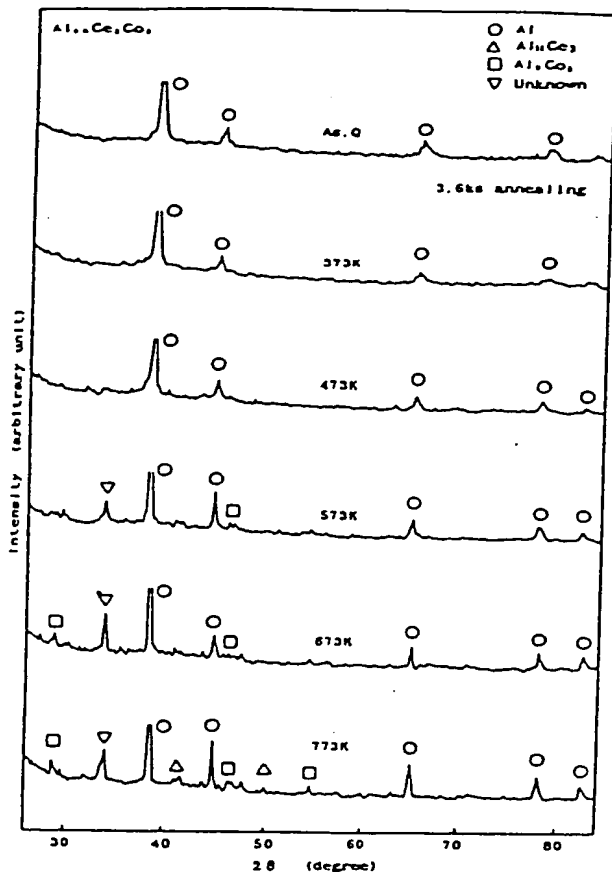


Fig. 8 X-ray diffraction patterns of $Al_{95}Ce_2Co_3$ alloy annealed for 3.6 ks at various temperatures.

ルファス単相領域では $Al_{86}Ce_4Co_{10}$ 合金の場合に最も高い硬度を示し、 H_v 375 である。しかし、全般的には Ce および Co 量の影響は明瞭には認められない。Fig. 6 および Fig. 7 より、fcc Al 単相領域では引張強度および硬度が共に高い合金は $Al_{97}Ce_2Co_1$ 合金であり、Al-Ce-Co 系 3 元合金においては、低溶質濃度の fcc Al 単相合金が熱的安定性や延性の観点か

らも望ましい合金であると考えられる。

3.6 焼なまし処理

上述したように fcc Al 単相領域の合金が望ましいことがわかったので、焼なまし処理は fcc Al 単相領域の合金について行った。Fig. 8 に $Al_{95}Ce_2Co_3$ 合金の急冷材および焼なまし材の X 線回折パターンを示す。図より 473 K までは 3.6 ks の焼なまし処理後も fcc Al 単相であることがわかる。573 K になると fcc Al 相から Al_9Co_2 化合物が析出する。さらに 773 K では $Al_{11}Ce_3$ 化合物が析出する。Fig. 9 に $Al_{95}Ce_3Co_2$ 合金の急冷材および焼なまし材の X 線回折パターンを示す。この場合も $Al_{95}Ce_2Co_3$ 合金の場合と同様に、473 K までは 3.6 ks の焼なまし処理によっても fcc Al 単相であるが、573 K では Al_9Co_2 化合物が析出し、さらに 673 K で $Al_{11}Ce_3$ 化合物を析出し、 $Al_{95}Ce_2Co_3$ 合金の場合よりも低温で $Al_{11}Ce_3$ 化合物が析出する。これは $Al_{95}Ce_3Co_2$ 合金が $Al_{95}Ce_2Co_3$ 合金よりも不安定なアモルファス相が多いことが原因と考えられる。Table 1 に種々の

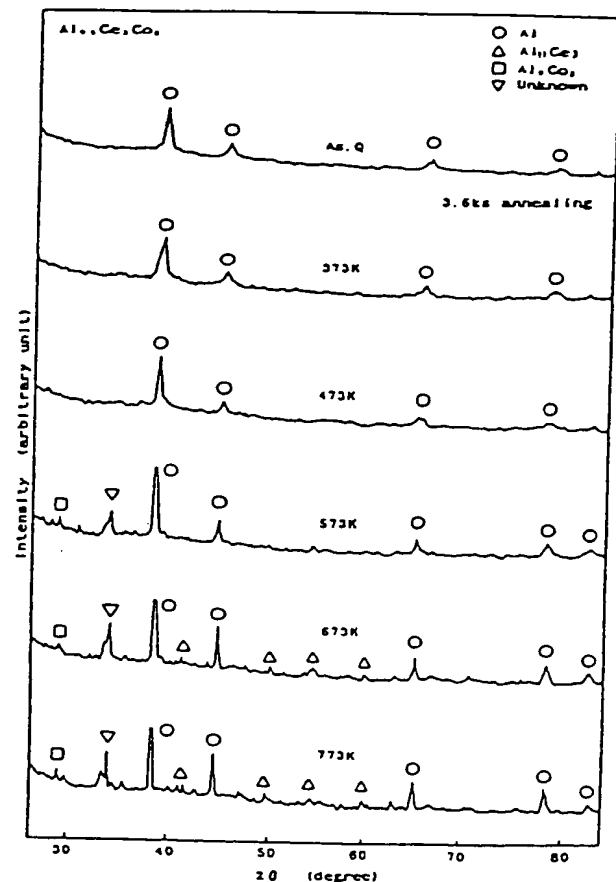


Fig. 9 X-ray diffraction patterns of $Al_{95}Ce_3Co_2$ alloy annealed for 3.6 ks at various temperatures.

そらく Al_9Co_2 化合物であると考えられる。化合物の析出場所については粒界のほかに粒内も考えられるが、本研究では明確に示すにはいたらなかった。Fig. 11 に $\text{Al}_{97}\text{Ce}_2\text{Co}_1$ 、 $\text{Al}_{95}\text{Ce}_2\text{Co}_3$ および $\text{Al}_{95}\text{Ce}_3\text{Co}_2$ 合金の焼なまし温度による fcc Al の格子定数の変化を示す。いずれの合金も一度減少してから増加する傾向を示している。格子定数の減少は格子間原子、原子空孔などが焼なましによって次第に減少することによると考えられる。 $\text{Al}_{95}\text{Ce}_2\text{Co}_3$ 合金の場合、急冷状態ではほとんど fcc Al 相であり、原子半径の大きい Ce が過飽和に固溶しているため格子定数が大きくなっていると考えられる。 $\text{Al}_{95}\text{Ce}_3\text{Co}_2$ 合金の場合には fcc Al 相とアモルファス相が共存しており、アモルファス相にも Ce が固溶しているため fcc Al の格子定数への Ce の影響が小さくなるためであると考えられる。合金によって格子定数が増加し始める温度に相違はあるが、いずれの合金も化合物の析出に伴って格子定数が変化している。Fig. 12 に $\text{Al}_{98}\text{Ce}_1\text{Co}_1$ 、 $\text{Al}_{96}\text{Ce}_1\text{Co}_3$ および $\text{Al}_{97}\text{Ce}_2\text{Co}_1$ 合金の焼なまし温度による引張強度 (σ_f) と硬度 (H_v) の変化を示す。 σ_f は低溶質濃度の $\text{Al}_{98}\text{Ce}_1\text{Co}_1$ 合金の場合焼なましの影響は小さいが、Ce 量や Co 量が増加すると σ_f の低下する温度は高温側に移行し、 $\text{Al}_{96}\text{Ce}_1\text{Co}_3$ および $\text{Al}_{97}\text{Ce}_2\text{Co}_1$ 合金は 573 K 付近から急激に σ_f は減少し、673 K 以上ではほぼ一定の値となり、 $\text{Al}_{98}\text{Ce}_1\text{Co}_1$ 合金とはほぼ同じ値となっている。これは Fig. 10 で示した透過電顕写真の結果からもわかるように、Ce 量や Co 量が多くなるにつれ、化合物が fcc Al 相の粒界に析出してくるためと考えられる。次に硬度について述べる。Ce 量も Co 量も少ない $\text{Al}_{98}\text{Ce}_1\text{Co}_1$ 合金の場合、焼なまし温度の上昇と共に H_v はゆるやかに減少しているが、 $\text{Al}_{96}\text{Ce}_1\text{Co}_3$ と $\text{Al}_{97}\text{Ce}_2\text{Co}_1$ 合金は 473 K 付近から減少している。これは化合物の析出によっても影響をうけるが、急冷状態における種々の欠陥の減少に伴い格子ひずみが徐々に除去されることによっても影響をうけるものと考えられる。低溶質濃度の fcc Al 単相合金の場合にはこれらの影響が少ないため、 H_v の変化の度合いが小さいと考えられる。Table 2 に Al-Ce-Co 合金の相領域別の焼なまし温度による密着曲げ性を示す。○印は延性を有する合金であり、●印は脆性を示した合金である。表より fcc Al 単相領域の合金は、773 K まではすべて延性を有していることがわかる。fcc Al 相とアモルファス相の共存領域の合金は $\text{Al}_{94}\text{Ce}_3\text{Co}_3$ と $\text{Al}_{94}\text{Ce}_4\text{Co}_2$ 合金以外は 373 K まではすべて延性を有し、 $\text{Al}_{94}\text{Ce}_3\text{Co}_1$ 合金

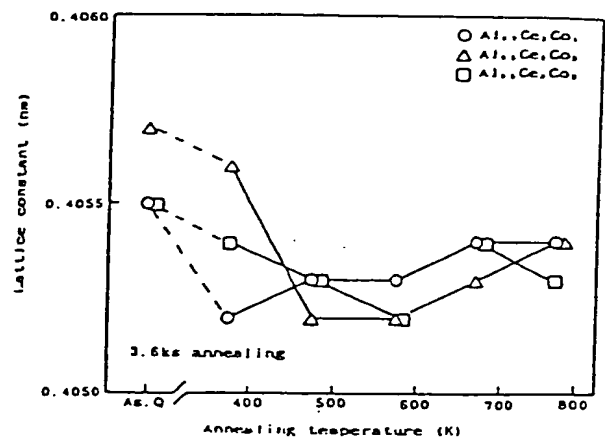


Fig. 11 Change in the lattice parameter of the fcc Al phase in rapidly solidified $\text{Al}_{97}\text{Ce}_2\text{Co}_1$, $\text{Al}_{95}\text{Ce}_2\text{Co}_3$ and $\text{Al}_{95}\text{Ce}_3\text{Co}_2$ alloys with annealing temperature.

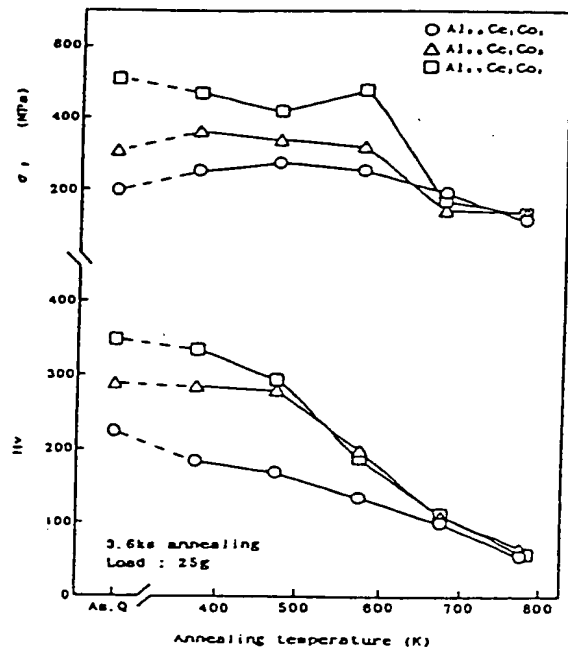


Fig. 12 Change in σ_f and H_v of rapidly solidified $\text{Al}_{98}\text{Ce}_1\text{Co}_1$, $\text{Al}_{96}\text{Ce}_1\text{Co}_3$ and $\text{Al}_{97}\text{Ce}_2\text{Co}_1$ alloys with annealing temperature.

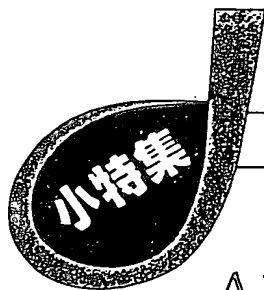
の場合には 773 K でも延性を示している。また $\text{Al}_{91}\text{Ce}_3\text{Co}_4$ 合金では 473 K でも延性を示している。以上のことから fcc Al 単相領域の合金では焼なましによって析出する化合物の延性への影響は小さいといえる。fcc Al 相とアモルファス相共存領域の合金では Co 量が少ない場合には焼なまし温度が高くても延性を有するが、Ce 量と Co 量が多くなると脆性を示すようになり、化合物の析出の影響が大きくなるものと考えられる。アモルファス単相領域の合金

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The translation of the C section

Fig. 5 shows $\sigma_{0.2}$ after holding alloy of Al₉₃Fe₃Cr₂Ti₂-26P/M material made of powder 26 micrometers or less, Al₉₃Fe₃Cr₂Ti₂-75P/M material and Al₉₂Fe₄Cr₂Ti₂-75P/M each made of powder 75 micrometers or less at the temperature of 300K, 473K, 573K and 673K for 100 hours. Fig.5 also shows $\sigma_{0.2}$ of Al₉₅Fe_{4.2}Ce_{0.8} alloy-P/M material which has the high heat resistance intensity produced by the P/M method and 2000 system Al - I/M material of marketing produced by dissolution casting method (Ingot metallurgy: I/M method). $\sigma_{0.2}$ of Al - I/M material falls rapidly from 400K, and is set to about 100 MPa by 573K. On the other hand, $\sigma_{0.2}$ of Al₉₅Fe_{4.2}Ce_{0.8} alloy P/M material shows high heat resistance intensity, and shows a maximum of 250 MPa at 573K. Moreover, $\sigma_{0.2}$ of this development Al alloy P/M material is high compare with Al alloy I/M material and Al-Fe-Ce alloy P/M material in all examination temperature.



Al 基および Zr 基合金のナノ準結晶分散組織の生成と高強度・高延性化

井上 明久* 木村 久道**

1. ま え が き

第2相をほとんど含まない準結晶単相は、Al, Mg, Pd, Zn, Ti 系合金⁽¹⁾の外に、最近では Zr⁽²⁾や Hf⁽³⁾系合金においても生成することが見出され、これらの準結晶合金の種々の基礎物性が調べられている⁽¹⁾。機械的性質に注目する時、準結晶単相は常温近傍では極めて硬くて⁽¹⁾⁽⁴⁾、塑性変形できず、脆い⁽⁴⁾ことが明らかにされている。また、準結晶単相のヤング率は極めて高く⁽⁵⁾、しかも熱膨張係数は小さい⁽⁵⁾⁽⁶⁾ことが報告されている。高い硬さとヤング率および小さい熱膨張係数から、準結晶を主体とする合金の脆さが克服される時、この種の合金が従来の結晶合金では得られない優れた機械的性質を示すことが期待される。

我々は、20~50 nm 径のほぼ球形のナノ粒径準結晶を結晶粒界を含まない fcc-Al 相⁽⁷⁾あるいは Zr 基⁽²⁾や Hf 基⁽³⁾金属ガラス相で取り囲んだナノ複相組織を生成することにより、準結晶の体積率が約80%の高い値まで、引張強度と粘さが増大することを見出している。これらのナノ準結晶粒子分散型の fcc-Al 合金および金属ガラスは全く新しいタイプの構造用高強度材料といえる。

本稿では、液体急冷法、銅鑄型鑄造法および粉末冶金法で作製した Al 基および Zr 基合金におけるナノ準結晶粒子分散型複相組織の生成とその原因および機械的性質について紹介する。

2. ナノ準結晶分散 Al 基合金

正20面体準結晶相は Mn⁽⁸⁾, Cr⁽⁹⁾, V⁽¹⁰⁾⁽¹¹⁾を溶質元素として含む液体急冷 Al 合金で生成することが知られている。これら合金の特徴である高い硬さとヤング率および高耐熱性の利用により、高強度耐熱 Al 合金の開発が期待される。その開発の鍵は準結晶を主相とする良好な延性をもつ合金が作製できるか否かにある。

図1は液体急冷 $Al_{99-x-y}Cr_xCe_1Co_y$ 4 元合金の曲げ延性と引張破断強度(σ_t)の組成依存性を示す⁽¹²⁾。密着曲げ変形

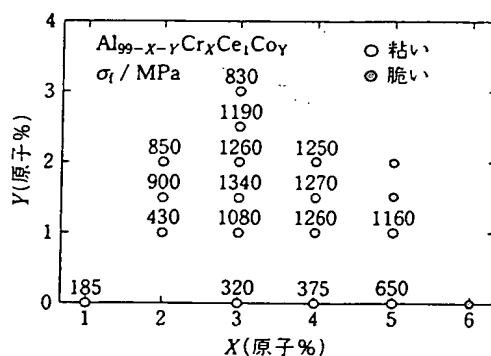


図1 液体急冷 $Al_{99-x-y}Cr_xCe_1Co_y$ 系 4 元合金の曲げ延性および引張破断強度(σ_t)の組成依存性。

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Formation and High Strength-Ductility Characteristics of Al- and Zr-based Alloys with Dispersed Nano-quasicrystalline Structures; Akihisa Inoue, Hisamichi Kimura (Institute for Materials Research, Tohoku University, Sendai)

Keywords: quasicrystal, aluminum-based alloy, zirconium-based alloy, nano-quasicrystalline particle, dispersed structure, high strength, good ductility

2000年5月1日受理

い硬さと熱的安定性をもつ準結晶粒子がP/M材中に分散していることは、これらP/M材が高強度、高耐熱強度および高耐摩耗性をもつことを示唆している。

表2は26 μm 以下($-26 \mu\text{m}$), 75 μm 以下($-75 \mu\text{m}$) およ

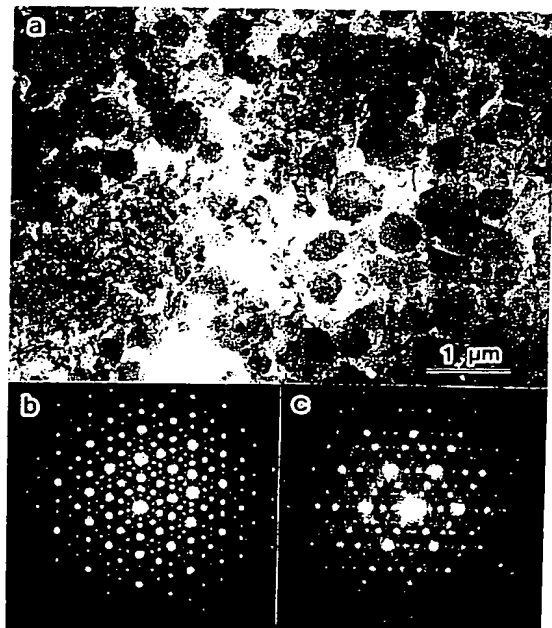


図4 押出し温度623 Kで作製した $\text{Al}_{94}\text{V}_4\text{Fe}_2$ 合金P/M材の透過電顕明視野像(a)と電子回折図形(b), (c)。

び125 μm 以下($-125 \mu\text{m}$)に篩い分けした粉末を用いて作製したAl-V系およびAl-Fe系合金P/M材の室温における最大引張強度(σ_{UTS}), 0.2%耐力($\sigma_{0.2}$), 伸び(ϵ_p), Hvおよび比強度[$\sigma_{\text{UTS}}/\text{密度}(\rho)$]をまとめている。表中には比較材として、超々ジュラルミン(7075-T6合金)の結果も併せて示している。表に見るように、Al-Fe-Cr-Ti系合金P/M材は7075-T6合金を越える高強度特性を有している。図5は26 μm 以下の粉末を用いて作製した $\text{Al}_{93}\text{Fe}_3\text{Cr}_2\text{Ti}_2$ 合金P/M材($\text{Al}_{93}\text{Fe}_3\text{Cr}_2\text{Ti}_2$ -26P/M材), 75 μm 以下の粉末の

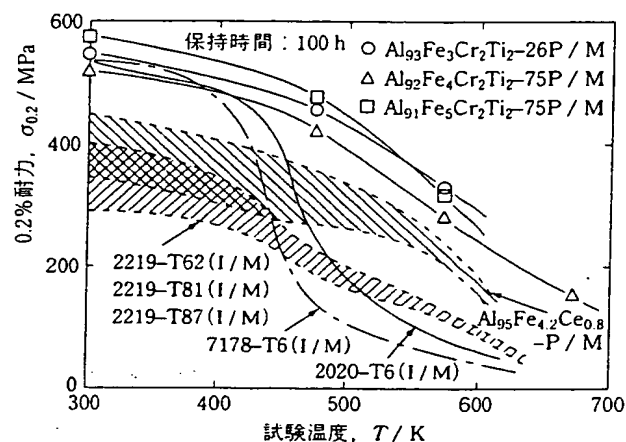


図5 Al-Fe-Cr-Ti系合金P/M材を試験温度300 K, 473 K, 573 K, 673 Kに100時間保持後の0.2%耐力($\sigma_{0.2}$)。

表1 エネルギー分散型X線分析法(TEM/EDX)で測定したAl-V系およびAl-Fe系合金P/M材の準結晶粒子の組成。(原子%)

合金/原子%	Al	Fe	Cr	Ti	V	Mn
$\text{Al}_{94}\text{V}_4\text{Fe}_2$	84.2 ± 2.9	3.8 ± 0.5	—	—	12.0 ± 2.7	—
$\text{Al}_{93}\text{Fe}_3\text{Cr}_2\text{Ti}_2$	84.6 ± 1.4	7.0 ± 0.8	6.0 ± 0.7	2.4 ± 0.7	—	—
$\text{Al}_{93}\text{Cr}_3\text{Fe}_2\text{Ti}_2$	84.6 ± 1.3	4.5 ± 0.9	8.2 ± 1.1	2.7 ± 0.7	—	—
$\text{Al}_{93}\text{Fe}_3\text{Cr}_2\text{V}_2$	83.0 ± 1.2	5.9 ± 1.1	5.2 ± 0.8	—	5.9 ± 0.4	—
$\text{Al}_{93}\text{Fe}_3\text{Mn}_2\text{Ti}_2$	82.9 ± 0.3	9.0 ± 0.5	—	1.8 ± 0.6	—	6.3 ± 0.2

表2 Al-V系およびAl-Fe系合金P/M材の最大引張強度(σ_{UTS}), 0.2%耐力($\sigma_{0.2}$), 伸び(ϵ_p), ビッカース硬さ(Hv)および比強度(最大引張強度/密度(ρ))。

合金/原子%	最大引張強度 $\sigma_{\text{UTS}}/\text{MPa}$	0.2%耐力 $\sigma_{0.2}/\text{MPa}$	伸び $\epsilon_p/\%$	ヤング率 E/GPa	ビッカース硬さ Hv	比強度(σ_{UTS}/ρ) $/10^5 \text{ Nm} \cdot \text{kg}^{-1}$
$\text{Al}_{94}\text{V}_4\text{Fe}_2$	585	490	4.5	85	190 ± 2	1.91
$\text{Al}_{93}\text{Fe}_3\text{Cr}_2\text{Ti}_2$	658 ± 9	545 ± 14	4.4 ± 0.5	85 ± 2	192 ± 3	2.20
$\text{Al}_{93}\text{Fe}_3\text{Mn}_2\text{Ti}_2$	608	490	4.2	87	190 ± 3	2.00
$\text{Al}_{93}\text{Fe}_3\text{Cr}_2\text{V}_2$	495 ± 2	370 ± 7	3.3 ± 0.2	86 ± 1	153 ± 2	1.70
$\text{Al}_{93}\text{Fe}_2\text{Cr}_3\text{Ti}_2$	608	550	3.5	91	188 ± 3	2.10
7075-T6	600	550	10.7	70	190 ± 3	2.10

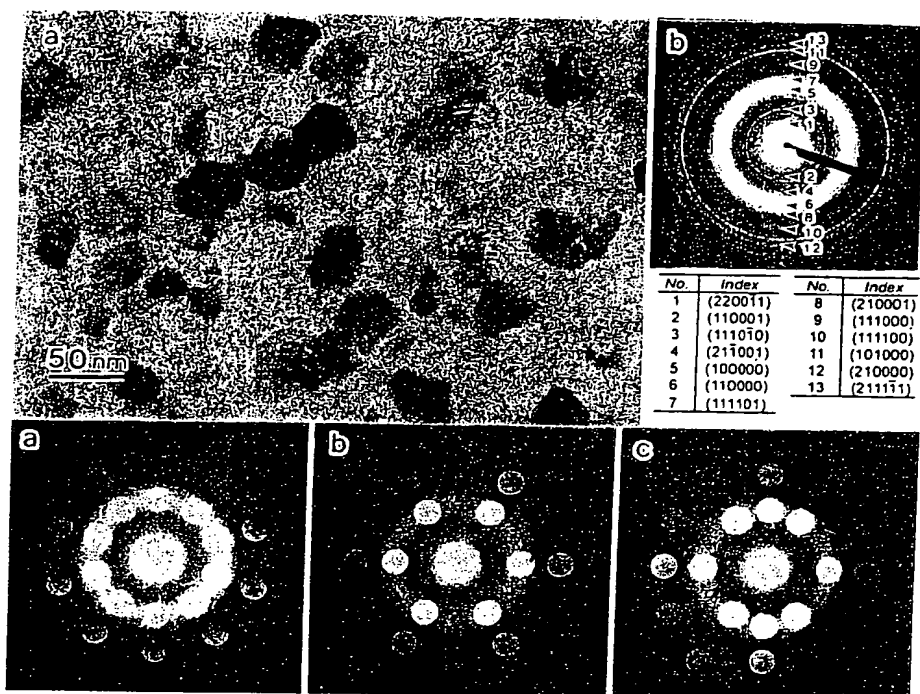


図8 705 Kで、60 s熱処理した $Zr_{65}Al_{7.5}Ni_{10}Cu_{7.5}Pd_{10}$ 金属ガラスの透過電顕明視野像(a)、電子回折図形(b)とナノビーム電子回折図形(c), (d), (e)。

Ptの場合には Zr_2Cu , Zr_2Ni , Zr_2Al_3 , $ZrPt$ であり、準結晶の残存は認められない。この結果は、これらの合金系で析出する準結晶は準安定相であるといえる。一例として、第一発熱ピーク直上まで加熱した $Zr_{65}Al_{7.5}Ni_{10}Cu_{7.5}Pd_{10}$ 合金のTEM明視野像、制限視野電子回折図形およびナノビーム電子回折図形を図8に示す。20~30 nm径のほぼ球形の析出物から得たナノビーム回折図形は、5回、3回、2回対称性を示しており、析出物は準結晶であると同定される。準結晶粒子は均一に析出し、約10 nm厚さの残存ガラス相で囲まれている。準結晶析出物の体積分率は70~80%であると推定される。

図8に示したZr基合金の準結晶+ガラス相組織は、図2に示したAl基合金の準結晶+fcc-Al相組織の特徴と類似している。すなわち、準結晶はナノ粒径の球形態であり、ガラス相あるいはAl相で囲まれた孤立状態にあり、 V_f は70~80%であり、母相には粒界は見られない。この類似した組織の特徴は、Zr基合金においてもAl基合金と同様な高強度、高延性特性が得られることが期待される。図9は銅鋳型鋳造法で作製したナノ準結晶分散 $Zr_{65}Al_{7.5}Ni_{10}Cu_{7.5}Pd_{10}$ バルクガラス材の圧縮応力-ひずみ曲線を示す⁽³¹⁾。また、図中には鋳造法で作製したバルクガラス合金のデータも比較のために示している。準結晶分散合金の σ_y , σ_t および ϵ_t はそれぞれ1780 MPa, 1830 MPaおよび3.1%であり、ガラス単相材での1510 MPa, 1640 MPaおよび2.2%に比べて、全ての特性において優れている。準結晶分散合金の破壊は、図10に示すように、応力軸に対して約45度傾いた最大剪断応力面上で生じ、破面には発達した脈状模様が見られる。この破面の特徴をガラス単相合金と比較する時、脈状模様はより細かいが、各脈の径はより大きくなっている。上記した剪断様式

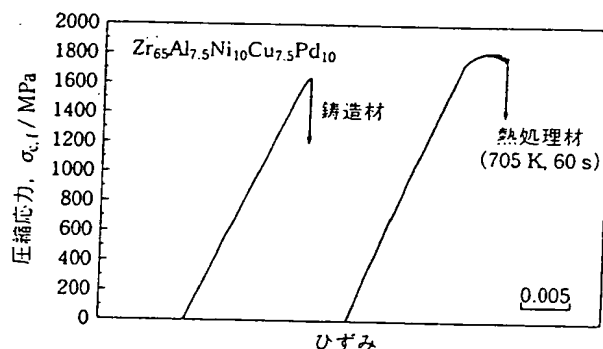


図9 $Zr_{65}Al_{7.5}Ni_{10}Cu_{7.5}Pd_{10}$ 金属ガラスの圧縮応力-ひずみ曲線。

と破面形態から、ナノ準結晶分散ガラス合金の変形と破壊は、図11に模式図的に示すように、最大剪断応力面に沿った粒間ガラス相内で局所化して生じているといえる。これらの変形、破断様式に基づく時、ナノ準結晶分散による強度と延性の増大は、(1)ナノ準結晶粒子がガラス相の剪断変形に対する有効な抑止体として働くこと、(2)変形が残存ガラス相に局所化することにより、ガラス相中の応力状態が変形能の増大をもたらす多軸応力状態になること、および(3)変形がガラス相に集中することにより局部的に温度が上昇し、変形能が増大すること、の要因の相乗効果に起因すると考えられる。

同様な強度と延性の増大は、引張応力条件下においても認められる。図12は銅鋳型鋳造法で作製したバルク形状の $Zr_{65}Al_{7.5}Ni_{10}Cu_{12.5}Ag_5$ ガラス合金の引張破断応力(σ_t)、引張破断伸び(ϵ_t)、ヤング率(E)およびHvの準結晶粒の V_f による変化をまとめている⁽³²⁾。 σ_t と ϵ_t は V_f が40%までの範

は小さい場合に限られている。ガラス構造には、正4面体、正8面体、正方12面体などの短範囲クラスターの存在が指摘されている⁽⁴²⁾。また、Zr-Al-Niなどの大きなガラス形成能と広い過冷却液体域を持つガラス合金の構造は、従来の 10^5 K/s以上の超急冷速度で作製されるアモルファス合金に比べてより高稠密な無秩序充填構造をもつと共に、その局所原子配列が対応する平衡結晶相とは異なった新しい局所構造を有していること、および構成元素は互いに引力相互作用を持ち、より長範囲にわたって均質な配列状態にあることが報告されている⁽²⁷⁾⁻⁽²⁹⁾。これらの特徴は、Zr-Al-Ni系合金のガラス構造が正20面体的な擬似分子構造⁽⁴³⁾を有していると仮定することと矛盾していない。この20面体的短範囲クラスターとM元素との関係を明らかにすることが、Zr-Al-(Ni, Cu)-M系での準結晶がガラス相からナノ粒子として $V_f=100\%$ の状態での析出できる原因を明らかにする上で重要と考えられる。さらに最近、Zr-Pd⁽⁴⁴⁾およびZr-Pt⁽⁴⁵⁾系2元合金においてもガラス相から正20面体準結晶が析出することが見出されており、この2元系準結晶がZr多成分系準結晶の基本であるとみなすことも出来、今後の解明が急がれる。

4. お わ り に

準結晶単相はきわめて硬くて脆いために、機械的性質を利用した構造用材料としての使用はほぼ絶望視されていた。しかし、球形のナノ粒徑準結晶を無粒界のfcc-Alあるいはガラス相で囲み、孤立粒子化を図ることにより、準結晶を主相とする合金においても高強度、高延性な特性が得られることが明らかになった。この成果は、準結晶の種々の特徴を反映した構造材料を開発できる可能性を示している。準結晶粒子のさらなる微細化および取り囲む周囲の相の種類や体積分率などを変化させることにより、さらに優れた特性のナノ準結晶粒子分散型高強度材料を開発できることが期待される。今後、この種の準結晶研究が益々活発化し、準結晶の実用化が果たされることを念願している。

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